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Richard B. Taylor

Date: May 2, 2008

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re application of)
)
EDWARD R. EATON ;)
WYNDHAM H. BOON; and)
CHRISTOPHER J. SMITH)
)
Serial No. 10/074,834)
)
Filed February 13, 2002)
)
A NOVEL CHEMICAL BASE FOR FUEL)
CELL ENGINE HEAT EXCHANGE)
COOLANT/ANTIFREEZE)
)
)
)

Group Art Unit: 1751

Examiner: Ogden Jr., N.

May 2, 2008

COMMISSIONER FOR PATENTS
P. O. Box 1450
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Sir:

APPEAL BRIEF

Please enter this appeal brief in the file of the above referenced patent application.

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(i) Real Party In Interest

The real party in interest of the present patent application is Shell Oil Company, the assignee of record.

(ii) Related Appeals and Interferences

There are no related appeals or interferences relative to the presently appealed application.

(iii) Status of Claims

Claims 1, 4, 5, and 6 are currently pending in the presently appealed patent application.

Claim 1 currently stands rejected under 35 U.S.C. §102(b) and 35 U.S.C. §103(a), and provisionally rejected on the grounds of non-statutory obviousness-type double patenting.

Claim 4 currently stands rejected under 35 U.S.C. §102(b) and 35 U.S.C. §103(a), and provisionally rejected on the grounds of non-statutory obviousness-type double patenting.

Claim 5 currently stands rejected under 35 U.S.C. §102(b) and 35 U.S.C. §103(a), and provisionally rejected on the grounds of non-statutory obviousness-type double patenting.

Claim 6 currently stands rejected under 35 U.S.C. §102(b) and 35 U.S.C. §103(a), and provisionally rejected on the grounds of non-statutory obviousness-type double patenting.

(iv) Status of Amendments

No amendment has been filed in the presently appealed patent application subsequent to the rejection that forms the basis of this appeal.

(v) Summary of the Claimed Subject Matter

Claims 1 and 4-6 are presently pending in the application and are involved in the appeal. One independent claim, claim 1, is involved in the appeal. Separate arguments are presented herein relative to dependent claims 4 and 5.

Claim 1 provides a nontoxic fuel cell engine coolant (supported in the original application text as filed at: p. 2, line 28; p. 4, lines 23-24; and original claim 1—the original application text is attached hereto as Exhibit 1). The coolant is comprised of an

aqueous 1,3-propanediol (supported in the original application text as filed at p. 3, lines 1-4; p. 14, lines 14-17 in combination with Figures 1 and 2; and original claims 3-5). The coolant has an electrical resistivity of greater than 250 Kohm-cm (supported in the original application text as filed at p.2, lines 28-29, p. 14, lines 7-13, and original claim 1). The coolant also has a boiling point of greater than 90°C, a thermal conductivity of greater than 0.4 W/m-k, a viscosity of less than 1 cPs at 80°C and of less than 0.6 cPs at 0°C, and a heat capacity of greater than 3 kJ/kg-K (supported in the original application text as filed at p. 2 lines 29-34 and in the original claim 1). Finally, the coolant has a corrosion of heat rejecting surface capacity as measured by ASTM D-4340 of less than 0.1 mg/cm²/week (supported in the original application text as filed in Table 2, p. 10, third row, and in Example 3).

Claim 4 provides the coolant of claim 1 wherein the aqueous 1,3-propanediol is comprised of from 40 to 85% by volume of 1,3-propanediol (supported in the original application text as filed at p. 3, lines 2-3).

Claim 5 depends from claim 4 and provides the coolant of claim 4 wherein the aqueous 1,3-propanediol is comprised of from 55 to 85% by volume of 1,3-propanediol (supported in the original application text as filed at p. 3, lines 2-3; and Example 4).

Claim 6 depends from claim 1 and provides that the coolant has a freezing point of less than -40°C (supported in the original application text as filed at p. 2, lines 31-32; p. 5, lines 24-26, Fig. 2, and original claim 6).

(vi) Grounds of Rejection to be Reviewed on Appeal

A. Whether claims 1 and 4-6 are unpatentable under 35 U.S.C. §102(b) as anticipated by Arntz et al. (U.S. Patent No. 5,015,789).

B. Whether claims 4 and 5 are unpatentable under 35 U.S.C. §102(b) as anticipated by Arntz et al. (U.S. Patent No. 5,015,789).

C. Whether claims 1 and 6 are unpatentable under 35 U.S.C. §102(b) as anticipated by JP 06026979.

D. Whether claims 1 and 4-6 are unpatentable under 35 U.S.C. §103(a) as

obvious over Artnz et al (U.S. Patent No. 5,015,789).

E. Whether claims 4 and 5 are unpatentable under 35 U.S.C. §103(a) as obvious over Artnz et al. (U.S. Patent No. 5,015,789).

F. Whether claims 1 and 4-6 are unpatentable under 35 U.S.C. §103(a) as obvious over JP 06026979.

G. Whether claims 1 and 4-6 are unpatentable on the ground of nonstatutory obviousness-type double patenting over claims of copending Application No. 10/886,298.

(vii) Argument

Legal Standards

The pertinent legal standards with respect to arguments presented in this application are provided below.

Anticipation

One of the requirements for patentability is that the claimed invention be new or novel. Thus, 35 U.S.C. § 102(b) of the Patent Statute provides that an applicant is not entitled to a patent if:

- (b) the invention was patented or described in a printed publication in this or a foreign country or in public use or on sale in this country, more than one year prior to the date of the application for patent in the United States...

35 U.S.C. §102 (2000). Thus, any patent or printed publication dated more than one year prior to the effective filing date of the application date of the patent at issue is considered prior art that may render a claim invalid.

A prior art reference anticipates a patent claim if that single reference discloses, either expressly or inherently, all limitations of the claim. *Finnigan Corp. v. Int'l Trade Comm'n*, 180 F.3d 1354, 1365 (Fed. Cir. 1999). Where a patent claim defines an element in terms of a range, any prior art disclosure containing a limitation that falls within the range will anticipate. *Atlas Powder Co. v. Ireco Inc.*, 190 F.3d 1342, 1346 (Fed. Cir. 1999). However, an anticipatory reference need not duplicate word for word what is set forth in the claims.

Anticipation occurs when a claimed limitation is inherent or otherwise implicit in the relevant reference. *Tyler Refrigeration v. Kysor Indus. Corp.*, 777 F.2d 687, 689 (Fed. Cir. 1985). Thus, a prior art reference without express reference to a claim limitation may nonetheless anticipate by inherency. See *In re Cruciferous Sprout Litig.*, 301 F.3d 1343, 1349 (Fed.Cir.2002). "Under the principles of inherency, if the prior art necessarily functions in accordance with, or includes, the claim's limitations, it anticipates." *Id.* (quoting *MEHL/Biophile Int'l Corp. v. Milgraum*, 192 F.3d 1362, 1365 (Fed.Cir.1999)). Inherency may not be established by probabilities or possibilities—the allegedly inherent characteristic must necessarily flow from the teaching of the prior art reference. See *In re Robertson*, 169 F.3d 743, 745; 49 U.S.P.Q.2d 1949, 1950-51 (Fed. Cir. 1999); and *Ex parte Levy*, 17 U.S.P.Q.2d 1461, 1464 (Bd. Pat. App. & Inter. 1990). The fact that a certain result or characteristic may occur or be present in the prior art is not sufficient to establish the inherency of that result or characteristic. *In re Rijckaert*, 9 F.3d 1531, 1534; 28 U.S.P.Q.2d 1955, 1957 (Fed. Cir. 1993). When relying on a theory of inherency, the examiner must provide a basis in fact and/or technical reasoning to reasonably support the determination that the allegedly inherent characteristic necessarily flows from the teachings of the applied art. *Ex parte Levy*, 17 U.S.P.Q.2d 1461, 1464 (Bd. Pat. App. & Inter. 1990).

Obviousness

The Patent Laws also require that an invention be "non-obvious". See 35 U.S.C. § 103. Non-obviousness is distinct from novelty in that an invention may be obvious even though it is not identically disclosed anywhere in the prior art. Thus, 35 U.S.C. § 103(a) of the patent statute provides that:

- (a) A patent may not be obtained though the invention is not identically disclosed or described as set forth in section 102 of this title, if the differences between the subject matter sought to be patented and the prior art are such that the subject matter as a whole would have been obvious at the time of the invention was made to a person having ordinary skill in the art to which said subject matter pertains.

35 U.S.C. § 103 (2000). Anticipation, however, is the ultimate or epitome of obviousness. *In re Grose et al.*, 592 F.2d 1161, 1165 (CCPA 1979). Everything which qualifies as prior art under Section 102(b) also qualifies as prior art under Section 103 as well.

The standard for proving obviousness requires that the invention must have been obvious to one of ordinary skill in the relevant art in light of the teachings of the prior art. *Ryko Mfg. Co. v. Nu-Star, Inc.*, 950 F.2d 714, 717 (Fed. Cir. 1991). The person of ordinary skill is a hypothetical person who is presumed to be aware of all the pertinent prior art. *Custom Accessories, Inc. v. Jeffery-Allan Indus., Inc.*, 807 F.2d 955, 962 (Fed. Cir. 1986). Obviousness is a question of law, based on a factual determination of the obviousness factors, discussed below. *Gambro Lundia AB v. Baxter Healthcare Corp.*, 110 F.3d 1573, 1576 (Fed. Cir. 1997).

To determine whether a patent is obvious, a court must determine: (1) the scope and content of the prior art; (2) the differences between the claimed invention and the prior art; (3) the level of ordinary skill in the relevant art; and, where applicable (4) objective or secondary considerations tending to prove non-obviousness. *See Graham v. John Deere*, 383 U.S. 1, 17-18 (1966); *Richardson-Vicks Inc. v. Upjohn Co.*, 122 F.3d 1476, 1479 (Fed. Cir. 1997). Obviousness generally involves the combining of one or more prior art references. *B. F. Goodrich Co. v. Aircraft Braking Sys. Corp.*, 72 F.3d 1577, 1582 (Fed. Cir. 1996)(a determination of obviousness may be based upon the teachings of a single prior art reference). However, there must be "some objective teaching in the prior art or that knowledge generally available to one of ordinary skill in the art would lead that individual to combine the relevant teachings of the references." *In re Fine*, 837 F.2d 1071, 1074 (Fed. Cir. 1988). The suggestion to combine references need not be explicit; it may be implicit. *In re Rouffet*, 149 F.3d 1350, 1355-56 (Fed. Cir. 1998); *B.F. Goodrich*, 72 F.3d at 1582.

Objective evidence tending to show that the invention was not obvious should also be considered and includes the following: long-felt need; failure of others; commercial success; industry acclaim and recognition; and licensing by industry. *Graham*, 383 U.S. at 17-18.

The initial burden of providing factual evidence supporting a *prima facie* conclusion of obviousness is borne by the examiner. MPEP 2142. An analysis supporting a rejection under 35 U.S.C. §103 should be made explicit. *KSR International Co. v. Teleflex, Inc.*, 82 U.S.P.Q.2d 1385, 1396 (2007). The key to supporting a rejection under 35 U.S.C. §103 is the clear articulation of the reason(s) why the claimed invention would have been obvious—there must be some articulated reasoning with some rational underpinning to support the legal conclusion of obviousness. *KSR International Co. v. Teleflex*, 82 USPQ 2d 1385, 1396 (2007). If the examiner does not establish a *prima facie* case of obviousness, the applicant is under no obligation to submit evidence of nonobviousness, but the burden of coming forward with evidence or arguments shifts to the applicant once the examiner has established a *prima facie* case of obviousness. MPEP 2142.

A. Whether claims 1 and 4-6 are unpatentable under 35 U.S.C. §102(b) as anticipated by Arntz et al (U.S. Patent No. 5,015,789—hereafter the "'789 patent").

1. The '789 patent does not expressly disclose all of the limitations of claim 1, and, as a result, does not expressly anticipate claim 1 and its dependent claims 4-6.

Claim 1 is set forth above, and is directed to a fuel cell engine coolant composition that is comprised of an aqueous 1,3-propanediol (hereinafter "PDO") and that has, among other physical characteristics, an electrical resistivity of greater than 250 Kohm-cm.

The '789 patent is directed to a method of hydrating acrolein to produce 3-hydroxypropionaldehyde (hereinafter "HPA"), and then hydrogenating HPA to produce PDO. The '789 patent discloses supplying a mixture of acrolein and water in the presence of an acidic cation exchanger resin containing acidic phosphonic acid groups to hydrate the acrolein to HPA. Unreacted acrolein is removed from the HPA containing solution, which is then hydrogenated to produce PDO. If the hydrogenation is conducted in a liquid phase, PDO may then be distilled from the resulting product mixture.

Claim 1 of the '789 patent is relied on to allegedly show that the current claims are expressly anticipated by the '789 patent. Claim 1 of the '789 patent provides:

A method of preparing 1,3-propanediol comprising hydrating acrolein in the presence of an acidic cation exchanger resin to form 3-hydroxypropionaldehyde, by reacting acrolein and water in a weight ratio of 1:2 to 1:20 at 30°C to 120°C and a pressure in a range from 1 to 20 bars in the presence of a cation exchanger resin having phosphonic acid groups, or salts thereof, separating said ion exchanger from the resulting reaction mixture and, to the extent it is present, unreacted acrolein to obtain 3-hydroxypropionaldehyde and subsequently catalytically hydrogenating the 3-hydroxypropionaldehyde in the liquid or gaseous phase using a hydrogenation catalyst therefor.

In the Office Action of September 14, 2007 the Examiner alleged that the '789 patent, in particular claim 1, disclosed all that was necessary to anticipate claims 1 and 4-6 of the present application. As acknowledged in the Office Action, however, the '789 patent is silent with respect to several of the limitations of the coolant claimed in claim 1 of the present application, including that the coolant has an electrical resistivity of greater than 250 Kohm-cm. As such, the '789 patent does not expressly anticipate claim 1 and its dependent claims 4-6 since the '789 patent does not expressly disclose any of the following limitations of the coolant claimed in claim 1 of the present application: an electrical resistivity of greater than 250 Kohm-cm; a boiling point of greater than 90°C; a thermal conductivity of greater than 0.4 W/m-k; a viscosity of less than 1 cPs at 80°C and less than 6 cPs at 0°C, a heat capacity of greater than 3kJ/kg-K, and a corrosion of aluminum heat rejecting surface capacity as measured by ASTM D-4340 of less than 0.1 mg/cm²/week.

2. The '789 patent does not inherently anticipate claims 1 and 4-6 of the present application.

In the Office Action of September 14, 2007, the Examiner alleged that, in the alternative to being expressly anticipated by the '789 patent, claim 1 and its dependent claims 4-6 were inherently anticipated by the '789 patent because the '789 patent discloses the same compound as claimed in claim 1 of the present application, and one of ordinary skill in the art would expect the compound disclosed in the '789 patent to encompass the claimed characteristics. In particular, the Examiner alleged that, as claimed, the coolant composition of claim 1 may only require aqueous PDO, and that the '789 patent provides an aqueous PDO product, and, therefore, claim 1 is inherently

disclosed in the '789 patent. This allegation is apparently based on the assumption that all aqueous PDO solutions inherently have an electrical resistivity of greater than 250 Kohm-cm.

Claim 1 and its dependent claims 4-6, however, are not inherently anticipated by the disclosure of the '789 patent, at least in part because the compositions provided by the '789 patent do not inherently provide an electrical resistivity of at least 250 Kohm-cm. Applicants note that, even if the claimed coolant composition consisted only of aqueous PDO, the claimed coolant composition would not be inherently disclosed by the '789 patent since the claimed electrical resistivity is not inherent to all aqueous PDO solutions. Therefore, merely because the '789 patent discloses an aqueous PDO solution does not mean that the '789 patent provides a composition that inherently provides an electrical resistivity of greater than 250 Kohm-cm.

Aqueous PDO, by definition, is comprised of PDO and water. As shown in Exhibit 2, water is known to have varying degrees of electrical conductivity (the inverse of resistivity) depending on the concentration of ions in the water. All naturally occurring water contains dissolved mineral salts that are separated into positively charged cations and negatively charged anions. Certain processes may be utilized to decrease the ionic content and increase resistivity of the water by removing ions from the water, for example reverse osmosis in which water is passed through a membrane to separate ions from the water, and deionization in which water is passed first through a cationic ion exchange resin to remove metal ions and subsequently through an anionic ion exchange resin to separate hydrogen ions released by the cationic ion exchange resin in the process of removing the metal ions from the water.

The conductivity/resistivity of the water present in an aqueous PDO solution affects the conductivity/resistivity of the aqueous PDO solution. As shown in the declaration of Glenn Komplin, attached as Exhibit 3, the electrical resistivity of aqueous PDO solutions varies depending on the amount of water in the aqueous PDO solution—where increased levels of water correlate to decreased electrical resistivity in an aqueous PDO system—and also vary according to the type of water in the solution—where, for example, aqueous PDO solutions formed from deionized water have much greater electrical resistivity than aqueous PDO solutions formed from tap water. The declaration

of Mr. Komplin further shows that the ionic content of the water of an aqueous PDO solution as well as the concentration of water in the aqueous PDO solution affects whether the aqueous PDO solution has an electrical resistivity of greater than 250 Kohm-cm.

It should be noted that the mixtures of water and PDO discussed in the Declaration are aqueous PDO mixtures since the Examiner indicated in the Office Action of September 14, 2007 that the claims of the present application do not specify any additional water employed in the coolant composition. No additional water is claimed to be utilized in the claimed coolant compositions, water + PDO = aqueous PDO, and the Declaration shows that not all aqueous PDO mixtures have the claimed electrical resistivity. Further, in the Office Action of September, 14, 2007, the Examiner stated that the showing of the Declaration of Mr. Komplin was not commensurate with the scope of the claimed invention for the showing of unexpected results—however, the Declaration is offered to show that all aqueous PDO solutions do not inherently have an electrical resistivity of greater than 250 Kohm-cm (and, therefore, a reference disclosing an aqueous PDO solution does not inherently anticipate claims 1, 4, and 5) rather than to show unexpected results.

The '789 patent makes no disclosure regarding the electrical resistivity of an aqueous PDO solution. The '789 patent merely provides a method of producing a product mixture that contains PDO and water from a mixture of acrolein and water. No disclosure is provided regarding the electrical resistivity or conductivity of the water used in the mixture of acrolein and water or the ionic content of the water used in the mixture—therefore, the water used in the mixture of acrolein may have a low electrical resistivity (moderately low to high ionic content), e.g. below 250 Kohm-cm, or a high electrical resistivity (very low ionic content) e.g. greater than 250 Kohm-cm. As a result, the mixture of acrolein and water may have a low electrical resistivity, e.g. below 250 Kohm-cm.

The process of producing an aqueous PDO product disclosed in the '789 patent either does not change the electrical resistivity of the aqueous solution as it proceeds from an aqueous acrolein mixture to an aqueous PDO mixture, or it decreases the electrical resistivity of the aqueous solution. The aqueous mixture of acrolein is contacted with an

acidic cation exchange resin to hydrate the acrolein to form HPA. Hydration of acrolein by contact of acrolein with a cation exchange resin and water does not increase or decrease the ionic concentration of the aqueous solution. The resin interacts with acrolein by donating a hydrogen to the olefinic bond of the carbon atom adjacent the carboxyl group, while water donates a hydroxyl group to the terminal olefinic carbon atom and a hydrogen to the resin to replace the hydrogen donated from the resin to hydrate the acrolein. No change in ionic concentration is created from the hydration reaction since the resin merely exchanges a hydrogen ion for a hydrogen ion, and the acrolein molecule gains the constituents of water. The hydrogenation of HPA to PDO does not affect the ionic concentration of the aqueous reaction mixture. The primary reactions of hydrating acrolein and then hydrogenating the resulting HPA to produce PDO, therefore, do not change the ionic concentration of the initial aqueous mixture, and the electrical resistivity of the aqueous PDO product mixture depends on the initial ionic concentration of the water in the aqueous acrolein mixture.

As noted above, the '789 patent does not provide any disclosure of the ionic concentration of the water in the aqueous acrolein mixture, nor does the '789 patent provide disclosure of the source of the water (e.g. ultrapure water, DI water, tap water, drinking water, boiler water etc.) from which an ionic concentration and electrical resistivity of the mixture may be inferred, nor does the '789 patent provide any information regarding the electrical resistivity or electrical conductivity of the water/acrolein mixture. Further, as noted above, aqueous mixtures may have greatly varying electrical resistivities depending on the ionic content of the mixture, and particularly, upon the ionic content of the water in the mixture. Therefore, there is no basis in fact and/or technical reasoning to reasonably support the determination that the allegedly inherent characteristic of aqueous PDO of an electrical resistivity of greater than 250 Kohm-cm necessarily flows from the teachings of the '789 patent.

Furthermore, the process of the '789 patent may actually produce a decrease in the electrical resistivity of the aqueous solution in the course of producing an aqueous PDO, depending on the ionic content of the initial aqueous acrolein mixture. Conductivity and resistivity of solutions are affected by the concentration of ions; the mobility of ions; the oxidation state of the ions; and the temperature of the solution (higher temperature

increases ion mobility and therefore increases conductivity and decreases resistivity). The process of the '789 patent includes a step of contacting an aqueous mixture of acrolein with a cationic ion exchange resin, which would serve to exchange hydrogen ions from the resin with metal ions, if present, in the solution (i.e. the first step of a water deionization process). Due to their small size, hydrogen ions are more mobile than the metal ions they replace—so the electrical conductivity of the aqueous mixture may increase and the electrical resistivity may decrease upon contact with the cationic ion exchange resin as the ionic mobility of the mixture increases. The '789 process does not include a step of subsequently contacting the mixture with an anionic ion exchange resin (the second and final step of a water deionization process in which hydrogen ions are removed from water), therefore, the hydrogen ions remain in the mixture in the aqueous PDO product and affect its electrical resistivity.

In summary, the Examiner's contention that the aqueous PDO compound of the '789 patent encompasses the invention as claimed in claim 1 since claim 1 only requires a coolant containing aqueous PDO, and the aqueous PDO compound of the '789 patent inherently will have the claimed electrical resistivity is incorrect since aqueous PDO mixtures may have an electrical resistivity outside the scope of the claim. The Examiner has failed to provide a basis in fact and/or technical reasoning to reasonably support the determination that the allegedly inherent characteristic of aqueous PDO of an electrical resistivity of greater than 250 Kohm-cm necessarily flows from the teachings of the '789 patent. In particular, the Examiner has failed to address the evidence presented by Applicants that aqueous PDO mixtures may have an electrical resistivity of less than 250 Kohm-cm.

B. Whether claims 4 and 5 are unpatentable under 35 U.S.C. §102(b) as anticipated by Arntz et al (U.S. Patent No. 5,015,789)

Claims 4 and 5 and the '789 patent are described above.

In addition to being novel over the '789 patent for the reasons discussed above, claims 4 and 5 of the present application are novel over the disclosure of the '789 patent since the '789 patent does not provide a composition containing aqueous PDO having from 40 to 85% PDO by volume, or from 55 to 85% PDO by volume.

The '789 patent does not provide an express disclosure of the concentration of PDO in the product mixture, therefore, the '789 patent does not expressly anticipate claims 4 and 5. The '789 patent does provide several examples from which the volume of PDO in the final product may be determined for comparison with the claims.

In Example 8, the reaction solution contains a maximum of 11.6% PDO by volume before being worked up by distillation. This may be determined as follows: 1.49 moles of HPA in an aqueous solution of 750 ml are converted at 78% yield to PDO, yielding 1.16 moles of PDO in solution (1.49 moles HPA x 0.78). PDO has a molecular weight of 80 g/mol and a density of 1.06 g/ml, so the reaction solution contains 87.5 ml of PDO ((1.16 moles PDO x 80 g/mol)/1.06 g/ml) in a 750 ml solution. This provides an aqueous solution containing 11.6 vol.% PDO ((87.5 ml/750 ml) x 100).

In Example 9, the reaction solution contains a maximum of 11.9% PDO by volume before being worked up by rotary evaporation and distillation. This may be determined as follows: a solution of water and acrolein containing 18.3 wt.% acrolein contains 18.3 g of acrolein per 100 g of the solution, which correlates to 0.326 mol acrolein per 100 g of solution (acrolein molecular weight = 56 g/mol, 18.3 g/56 g/mol=mol of acrolein). 49% of the acrolein is converted to HPA, so the conversion yields 0.159 mol HPA (0.49 x 0.326) per 100 g of solution. Unreacted acrolein is removed from the solution, which is a maximum of 0.166 mol (49% conversion to HPA, maximum unconverted 51% acrolein—0.326 x 0.51=0.166), which correlates to a maximum removal of 9.29 g from the solution (0.166 mol x 56 g/mol), so that the solution contains a maximum of 0.159 mol HPA per 90.3 g of solution (100 g solution – 9.29 g of acrolein=90.3 g remaining solution). Hydrogenation converts 77% of the reacted acrolein (the HPA) to PDO, so there is 0.122 mol PDO per 90.3 g of solution (0.159 mol HPA x 0.77= 0.122 mol PDO). This is equivalent to 11.3 g PDO per 90.3 g of solution (0.122 mol PDO x 80 g/mol (PDO)), which corresponds to PDO volume% of 11.9 vol.% [(11.3 g PDO/1.06 g/mol PDO)=10.6 ml PDO; solution is water + PDO with minor amounts of HPA and side products so solution will have approximately 79 ml water ((90.3 g solution – 11.3 g PDO)/1.0 g/mol water)), and the PDO volume percent is (10.6 ml PDO/(10.6 ml PDO + 79 ml H₂O))x100]. In a similar manner, the volume% of

PDO may be calculated for Example 10 as 15.4 vol.% PDO and for Example 11 as 10.1 vol.% PDO.

As shown, none of the aqueous PDO products provided in the Examples of the '789 patent provides an aqueous PDO solution containing from 40 to 85 vol.% PDO or from 55 to 85 vol.% PDO as claimed in claims 4 and 5, but rather, the aqueous products produced therein have a much lower volume percent of PDO. Therefore, the '789 patent neither expressly nor inherently discloses the compositions as claimed in claims 4 and 5.

C. Whether claims 1 and 6 are unpatentable under 35 U.S.C. §102(b) as anticipated by JP 06026979 (hereinafter the "JP patent").

1. The JP patent does not expressly disclose all of the limitations of claim 1, and, as a result, does not expressly anticipate claims 1 and its dependent claim 6.

Claim 1 is set forth above, and is directed to a fuel cell engine coolant composition that is comprised of an aqueous 1,3-propanediol (hereinafter "PDO") and that has, among other physical characteristics, an electrical resistivity of greater than 250 Kohm-cm.

The JP patent is directed to a leak detecting liquid composition for a dual shell underground tank that comprises an antifreezing agent, water, a rust preventing agent, and a preservative. The antifreezing agent in the composition may be PDO. The rust preventing agent is a nitrate salt. The preservative includes, among other components, a quaternary ammonium salt.

In the Office Action of September 14, 2007, the Examiner alleged that the JP patent disclosed all that was necessary to anticipate claims 1 and 6 of the present application. As acknowledged in the Office Action, however, the JP patent (apparently mistakenly referred to as the Arntz '789 patent) is silent with respect to an electrical resistivity of greater than 250 Kohm-cm, a boiling point of greater than 90°C, a thermal conductivity of greater than 0.4 W/m-k, a viscosity of less than 1 cPs at 80°C and less than 6 cPs at 0°C, a heat capacity of greater than 3 KJ/kd-K; and a corrosion of aluminum heat rejecting surface capacity of less than 0.1 mg/cm²/week. Therefore, the JP patent cannot expressly anticipate claim 1 or its dependent claim 6 since it fails to disclose numerous limitations in the claims.

2. The JP patent does not inherently disclose all of the limitations of claim 1, and, as a result, does not expressly anticipate claims 1 and its dependent claim 6.

In the Office Action of September 14, 2007, the Examiner alleged that, in the alternative to being expressly anticipated by the JP patent, claim 1 and its dependent claim 6 were inherently disclosed by the JP patent since the compositions disclosed in the JP patent allegedly have the same composition as claimed in claims 1 and 6, and one of ordinary skill in the art would expect similar characteristics.

Claim 1 and its dependent claim 6, however, are not inherently anticipated by the disclosure of the JP patent, at least in part because the compositions disclosed in the JP patent do not inherently provide an electrical resistivity of at least 250 Kohm-cm. The compositions disclosed in the JP patent include water and may include PDO as an antifreeze, but all of the compositions disclosed in the JP patent include a nitrate salt rust preventing agent and a quaternary ammonium salt. Referring to the application text as filed (Exhibit 1) page 6 line 30 through page 7 line 2, ionic corrosion inhibitors are disqualified in the coolants of the present invention due to the electrical field of the fuel cells in which the coolants are to be used. Referring again to Exhibit 2, one skilled in the art would be aware that the electrical conductivity (and inversely the electrical resistivity) of an aqueous solution is dependent upon the concentration of ions in the solution, where increasing ionic concentration results in increasing electrical conductivity and decreasing resistivity. The aqueous compositions of the JP patent all include nitrate salts and quaternary ammonium salts which increase the ionic content of the compositions, thereby decreasing electrical resistivity. One skilled in the art would not expect such salt containing solutions to have a high electrical resistivity, e.g. greater than 250 Kohm-cm, due to the salts in the compositions. Therefore, there is no basis in fact and/or technical reasoning to reasonably support the determination that the allegedly inherent characteristic of the aqueous PDO of claim 1 of an electrical resistivity of greater than 250 Kohm-cm necessarily flows from the teachings of the JP patent.

D. Whether claims 1 and 4-6 are unpatentable under 35 U.S.C. §103(a) as obvious over Artanz et al (U.S. Patent No. 5,015,789).

The key to supporting a rejection under 35 U.S.C. §103 is the clear articulation of the reason(s) why the claimed invention would have been obvious—there must be some articulated reasoning with some rational underpinning to support the legal conclusion of obviousness. *KSR International Co. v. Teleflex*, 82 USPQ 2d 1385, 1396 (2007). Applicants allege that the Examiner has provided no articulated reasoning with some rational underpinning to support the legal conclusion of obviousness, and, therefore, has failed to establish a *prima facie* case of obviousness of claim 1 and its dependent claims 4-6.

The Examiner has alleged that the present invention of claim 1 of a coolant composition comprising aqueous PDO having, among other characteristics, an electrical resistivity of greater than 250 Kohm-cm and a corrosion of heat rejecting surface capacity of less than 0.1 mg/cm²/week is obvious from the '789 patent since the '789 patent provides an aqueous PDO composition formed by hydrogenating an aqueous solution of 3-hydroxypropionaldehyde. The '789 patent, however, would not direct one skilled in the art to a coolant composition of aqueous PDO having an electrical resistivity of greater than 250 Kohm-cm and a corrosion of heat rejecting surface capacity of less than 0.1 mg/cm²/week. First, as noted above and as admitted by the Examiner, the '789 patent provides no disclosure regarding the electrical resistivity or corrosion of heat rejecting surface capacity characteristics of the aqueous PDO produced according to disclosed process. Indeed, the '789 patent provides no information about the aqueous PDO product. Rather, the '789 patent teaches that the aqueous PDO solution is to be distilled to remove water from the PDO to prepare PDO (see e.g. '789 patent col. 7, lines 43-46, and col. 8, lines 19-23). As such, the '789 patent discloses that the aqueous PDO produced by the disclosed process is an intermediate that is further processed to obtain PDO, and one skilled in the art would be directed away from an aqueous PDO composition as claimed in claim 1 of any of its dependent claims 4-6 by the '789 patent.

Furthermore, even if the '789 patent can be read as teaching the production of an aqueous PDO as an end product rather than an intermediate, one skilled in the art would not be directed by the disclosure of the '789 patent to produce the aqueous PDO coolant composition of claim 1 since one skilled in the art would not be directed to produce an aqueous PDO having an electrical resistivity of greater than 250 Kohm-cm or a corrosion

of heat rejecting surface capacity of less than 0.1 mg/cm²/week. While it is known in the art that diols, e.g. ethylene glycol, are useful in coolant compositions, an aqueous diol composition having high electrical resistivity and low corrosivity—which maintains the high resistivity of the composition by limiting leaching of ions into the composition—is unexpected (see application text as filed (Exhibit 1) p. 14, lines 7-13, particularly lines 12-13). The '789 patent provides no disclosure that would lead one skilled in the art to prepare such a composition—the '789 patent merely teaches that aqueous PDO is formed by hydrogenating 3-hydroxypropionaldehyde, and that the aqueous PDO is stripped of water to prepare PDO.

Therefore, no case of *prima facie* obviousness of claim 1 and its dependent claims 4-6 over the '789 patent has been established. The conclusion of obviousness based on 1) the mere presence of an aqueous PDO that is an intermediate in the production of PDO; or 2) an assumption that the mere disclosure of an aqueous PDO renders the claimed aqueous PDO composition having highly specific electrical resistivity and anti-corrosive properties; lacks a rational underpinning sufficient to support the conclusion of obviousness.

E. Whether claims 4 and 5 are unpatentable under 35 U.S.C. §103(a) as obvious over Arntz et al. (U.S. Patent No. 5,015,789).

The Examiner has alleged that the present invention of claims 4 and 5 are obvious from the '789 patent since the '789 patent provides an aqueous PDO composition formed by hydrogenating an aqueous solution of 3-hydroxypropionaldehyde. Claim 4 provides the coolant composition of claim 1 comprising 40 to 85% PDO by volume, and claim 5 provides the coolant composition of claim 4 comprising from 55 to 85% PDO by volume.

Applicants maintain that no *prima facie* case of obviousness of claims 4 and 5 over the '789 patent has been established since no rational underpinning has been provided sufficient to support the conclusion of obviousness.

The '789 patent does not provide an explicit disclosure of an aqueous PDO composition containing more than 15.4 vol.% PDO, as calculated above for Examples 8-11. Further, the '789 patent provides an aqueous acrolein starting material containing a maximum of 33 wt.% acrolein that is converted to 3-hydroxypropionaldehyde, preferably

at a conversion rate of 30 to 80%, which is then hydrogenated to form an aqueous solution of PDO (see '789 patent col. 3 lines 16-18 and 38-50). Therefore, assuming 100% conversion of 3-hydroxypropionaldehyde to PDO, the maximum PDO concentration in the aqueous PDO solution formed according to the '789 process would be 26.4 wt.% which corresponds to 24.9 vol% PDO. Furthermore, one skilled in the art would avoid increasing the concentration of acrolein in the aqueous acrolein starting material to avoid forming an aqueous solution containing a high concentration of 3-hydroxypropionaldehyde since 3-hydroxypropionaldehyde would be more likely to form acetal side-products by reaction with itself at high concentrations during hydrogenation, which would reduce the ultimate yield of PDO from the process. Therefore, no rational underpinning has been provided on the basis of the '789 patent that would render claims 4 or 5 *prima facie* obvious from the disclosure of the '789 patent.

F. Whether claims 1 and 4-6 are unpatentable under 35 U.S.C. §103(a) as obvious over JP 06026979 (the JP patent).

The Examiner has alleged that the present claim 1 and its dependent claims 4-6 are obvious over the JP patent since one of ordinary skill in the art would expect similar characteristics as those claimed from the leak detecting liquid compositions disclosed in the JP patent, and since it would have been obvious to employ PDO in the amounts claimed as one skilled in the art would have been motivated to optimize the proportions of ingredients in the absence of evidence to the contrary.

Applicants maintain that no *prima facie* case of obviousness of claim 1 and its dependent claims 4-6 over the '789 patent has been established since no rational underpinning has been provided sufficient to support the conclusion of obviousness.

As noted above, the JP reference provides a composition containing PDO, water, a rust preventing agent and a preservative, where, the rust preventing agent is a nitrate salt and the preservative contains a quaternary ammonium salt. One skilled in the art would not find the invention as claimed in claim 1 obvious over the JP patent since the JP patent teaches that a rust preventing agent is necessary in the composition containing PDO and water, where the rust preventing agent is a nitrate salt. As noted above, the nitrate salt will increase the electrical conductivity of the composition and decrease the

electrical resistivity of the composition. One skilled in the art would not find the present invention as claimed in claim 1 obvious over the JP patent since the composition of claim 1 has a low degree of corrosivity while having a high degree of resistivity—the composition of the JP patent sacrifices electrical resistivity to improve rust resistance (low corrosivity) by utilizing a nitrate salt. Therefore, the JP patent teaches one skilled in the art to combine water, PDO, and a nitrate salt to provide a composition with low corrosivity, which teaches away from making a composition having a high electrical resistivity. The JP patent further teaches utilizing a quaternary ammonium salt in the composition, which would further decrease the electrical resistivity of the composition. Therefore, no rational underpinning has been provided sufficient to support a conclusion that claim 1, and its dependent claims 4-6, are *prima facie* obvious from the disclosure of the JP patent.

G. Whether claims 1 and 4-6 are unpatentable on the ground of nonstatutory obviousness-type double patenting over claims of copending Application No. 10/886,298.

The Examiner has alleged that claims 1 and 4-6 are unpatentable over copending Application No. 10/886,298 on the ground of non-statutory obviousness-type double patenting. Applicants are not the assignee of Application No. 10/886,298, which is assigned to Dupont-Staley BioProducts (now DuPont Tate & Lyle BioProducts, LLC), although the applications have a common inventor (Applicants, however, have asserted to DuPont-Staley BioProducts that they believe themselves to be a co-owner of Application No. 10/886,298 which is contested by DuPont-Staley BioProducts). As stated in the MPEP §804(I)(1): "[I]f a 'provisional' nonstatutory obviousness-type double patenting (ODP) rejection is the only rejection remaining in the earlier-filed of the two pending applications, while the later-filed application is rejectable on other grounds, the examiner should withdraw that rejection and permit the earlier-filed application to issue as a patent without a terminal disclaimer. If the 'provisional' ODP rejections in two applications are the only rejections remaining in those applications, the examiner should withdraw the ODP rejection in the earlier-filed application thereby permitting that application to issue without need of a terminal disclaimer."

The present application was filed on February 13, 2002, while, as shown by the cover sheet of the publication of Application No. 10/886,298 (attached as Exhibit 4), Application No. 10/886,298 was filed on July 6, 2004 and claimed priority from Provisional Application No. 60/485,441 filed July 7, 2003. The present application is, therefore, the earlier-filed of the two pending applications, including the provisional application from which Application No. 10/886,298 claims priority. Applicants maintain that the rejections under 35 USC §§ 102 and 103 should be obviated by the present appeal and should no longer apply, therefore, Applicants allege that the only remaining rejection should be the double patenting rejection. As such, as stated in the MPEP, that rejection should be withdrawn since the present application is the earlier-filed of the two patent applications.

Applicants would also draw the Patent Office's attention to the status of Application No. 10/886,298. On April 2, 2008 a Notice of Allowance was issued in the case of Application No. 10/886,298. No rejection was made in that application on the basis of obviousness-type double patenting over the present application. Applicants assert that if Application No. 10/886,298 is allowable over the claims of the present application with no obviousness-type double patenting rejection, then the claims of the present application are allowable over the claims of Application No. 10/886,298, and that the obviousness-type double patenting rejection is improper in the present case.

(viii) Claims Appendix

1. A nontoxic fuel cell engine coolant which is comprised of an aqueous 1,3-propanediol and which has an electrical resistivity of greater than 250 Kohm-cm, a boiling point of greater than 90°C, a thermal conductivity of greater than 0.4 W/m-k, a viscosity of less than 1 cPs at 80°C and less than 6 cPs at 0°C, a heat capacity of greater than 3 kJ/kg-K, and having a corrosion of aluminum heat rejecting surface capacity as measured by ASTM D-4340 of less than 0.1 mg/cm²/week.
4. The coolant of claim 1 wherein the solution is comprised of from 40 to 85% by volume of 1,3-propanediol.
5. The coolant of claim 4 wherein the solution is comprised of from 55 to 85% by volume of 1,3-propanediol.
6. The coolant of claim 1 having a freezing point of less than -40°C.

(ix) Evidence Appendix

Exhibit 1—Application text as filed. Entered into the record on February 13, 2002 along with the original filing documents. Copy attached.

Exhibit 2—Exhibit regarding Measurement of Conductivity. Entered into the record on July 19, 2006 as pages 7-10 of the Applicant Arguments/Remarks Made in an Amendment filed on July 17, 2006. Copy attached.

Exhibit 3—Declaration of Mr. Glenn Komplin under 37 CFR §1.132. Entered into the record on December 11, 2006. Copy attached.

Exhibit 4—Cover sheet of publication of Application No. 10/886,298. Copy attached.

(x) Related Proceedings Appendix

None.

Exhibit 1



A NOVEL CHEMICAL BASE FOR FUEL CELL ENGINE
HEAT EXCHANGE COOLANT/ANTIFREEZE

5

Field of the Invention

This invention relates to a novel technology for use in cooling systems for fuel cell powered vehicles and/or equipment. In order to remove the heat that is generated in fuel cell systems, 1,3-propane diol is used as the chemical base for the heat exchange fluid.

Background of the Invention

It has been suggested that fuel cell technology can be used to generate electricity in sufficient volume to be applicable in the driving of electric motors for passenger vehicles, standby power generation, and other applications. A fuel cell is a device that converts chemical energy of a fuel directly into electricity and they are intrinsically more efficient than most other energy generation devices, such as internal combustion engines. In principle, a fuel cell operates somewhat like a battery. Unlike a battery, a fuel cell does not run down or require recharging. It will produce energy in the form of electricity and heat as long as fuel is supplied. The most common type of fuel cell consists of two electrodes sandwiched around an electrolyte. Oxygen passes over one electrode and hydrogen over the other, generating electricity, water, and heat.

The fact that heat is generated by the fuel cell requires the presence in the automobile or other system of a cooling system which can be similar to those used presently in internal combustion engines. Typically, such a system includes a circulating pump, plumbing that may include aluminum, brass, copper, lead-tin solder, stainless steel, plastic or rubber materials, and a heat

exchanger (radiator) typically constructed of aluminum or copper/brass.

The heat exchange fluid (coolant) is obviously just as important in a fuel cell system as it is in internal combustion engines. Many of the requirements of a heat exchange fluid for internal combustion engines are also required for fuel cell engines. However, there are some additional requirements. For instance, fuel cell vehicles generate a direct current of 400 volts. The coolant, which flows around the aluminum components of the fuel cell, must be nonconductive to protect both the cell itself from shorting out and to prevent electrical hazard to humans operating or servicing the system.

The first fuel cell was built in 1839 by Sir William Grove, a Welsh judge and gentleman scientist. The "Grove cell" used a platinum electrode immersed in nitric acid and a zinc electrode in zinc sulfate to generate about 12 amps of current at about 1.8 volts. There were other developments in fuel cell technology over the years but serious interest in the fuel cell as a practical generator of electricity did not begin until the 1960's, when the U.S. Space Program chose fuel cell technology over nuclear power and solar energy. This technology, developed by Francis Thomas Bacon, used nickel gauze electrodes and operated under pressures as high as 300 psi.

Summary of the Invention

A nontoxic fuel cell engine coolant which has an electrical resistivity of greater than 250 kOhm-cm, a boiling point of greater than 90°C, optionally, a freezing point of less than -40°C, a thermal conductivity of greater than 0.4 W/m-k, a viscosity of less than .1 cPs at 80°C, a viscosity of less than 6 cPs at 0°C, a heat capacity of greater than 3 kJ/kg-K, and which is

compatible with current cooling system materials. The coolant may contain from 1 to 100, preferably 40 to 85 and most preferably 55 to 85, volume percent PDO and most or all of the remaining balance is water.

5 Brief Description of the Drawings

Figure 1 illustrates the aqueous solution freeze point characteristics of the 1,3-propanediol and GM 6043 inhibition chemistry (EG).

Figure 2 is a plot of the freeze behavior of aqueous 1,3-propanediol antifreeze.

10 Detailed Description of the Invention

As previously stated, the purpose of a fuel cell is to produce an electrical current that can be directed outside the cell to do work, such as powering an electric motor. Because of the way electricity behaves, this current returns to the fuel cell, completing an electrical circuit. The chemical reactions that produce this current are the key to how a fuel cell works. There are several kinds of fuel cells which operate somewhat differently but in general terms, hydrogen atoms enter a fuel cell at the anode where a chemical reaction strips them of their electrons. The hydrogen atoms are now "ionized" and carry a positive electrical charge. The negatively charged electrons provide the current through wires to do work.

Oxygen enters the fuel cell at the cathode and it there combines with electrons returning from the electrical circuit and hydrogen ions that have traveled through the electrolyte from the anode. In some fuel cells the oxygen picks up electrons and then travels through the electrolyte to the anode where it combines with hydrogen ions. This chemical reaction generates a significant amount of heat energy which must be removed

from the fuel cell in order for it to continue to operate properly.

A number of objectives have been identified for coolants for fuel cell vehicles. First, since fuel cell vehicles generate a direct current of 400 volts, the coolant which flows around the aluminum components of the fuel cell must be nonconductive to protect the cell from shorting out and to prevent electrical hazards. Other physical property objectives for fuel cell coolants are set out in the table below:

Table 1

Electrical Resistivity	>250 kOhm-cm
Boiling point	> 90°C
Freezing point	< -40°C
Thermal Conductivity	> 0.4 W/m-k
Viscosity	< 1 cPs @ 80°C < 6 cPs @ 0°C
Heat Capacity	> 3 kJ/kg-K
Durability	> 5,000 hours of operation/3 years total time
Material compatibility:	Compatible with current cooling system materials
Toxicity	Classified as non-toxic for transportation

1,3-propanediol (PDO), which is manufactured by Shell Chemical Company, is generally made as described in US patent No. 5,304,691 and the art described therein. This is a process for making PDO and HPA (3-hydroxypropanal, a 3-hydroxyaldehyde). In this particular patent, PDO and HPA are made by intimately contacting an oxirane (ethylene oxide, hereinafter 'EO'), a ditertiary phosphine-modified cobalt carbonyl catalyst, a ruthenium catalyst promoter, and syngas (carbon monoxide and hydrogen) in an inert reaction solvent at hydroformylation reaction conditions. A PDO yield of up to 86-87 mole% is reported, using a catalyst comprising cobalt ligated with 1,2-bis(9-phosphabicyclononyl)ethane

as bidentate ligand, and either triruthenium(0) dodecarbonyl or bis[ruthenium tricarbonyl dichloride] as cocatalyst. Other methods of making PDO are known.

Inhibited with the GM 6043 chemistry, the 1,3-propanediol performed somewhat better than EG in modified ASTM-type tests. Figure 1 illustrates the aqueous solution freeze point characteristics of the 1,3-propanediol and GM 6043 (EG). There is a slight compromise of freeze protection as determined by the ASTM D1177 test method, but the 1,3-propanediol was soft and slushy at the reported freeze point. This could be an indication that actual protection against hard, damaging freezing is actually better, approaching the effective protection point of the EG-based product. We also performed the D1177 test with 55% and 60% 1,3-propanediol in water, and found that the 55% concentrated product offered protection equivalent to 50% EG, per the test method. Freeze protection continued to improve at 60% 1,3-propanediol. We feel that the antifreeze properties of the chemistry are acceptable. Indeed a 50% solution would provide adequate protection against freezing in most geographies. TC in Figure 1 is an internal designation for the PDO aqueous solutions at 50, 55, and 60 volume percent PDO.

Figure 2 shows the freeze behavior of PDO/water solutions. It can be seen that formulations may be made with freeze points significantly lower than -40°C .

It may be desirable to include an effective amount of an antifoaming composition in the antifreeze/coolant composition. Such components are well known. Polyglycol-type antifoaming agents can be used.

PDO coolants in fuel cell vehicles will have an electrical resistivity of greater than 250 kOhm-cm, a boiling point of greater than 90°C , usually a freezing point of less than -40°C , a thermal conductivity of greater than

0.4 W/m-k, a viscosity of less than 1 cPs at 80°C and less than 6 cPs at 0°C, a heat capacity of greater than 3 kJ/kg-K, a desired durability of greater than 5000 hours of operation (three years total time), material compatibility - will not corrode or erode current automotive cooling system materials, have a toxicity classified as non-toxic for transportation, and will be cost competitive with current automotive coolants.

The PDO formulations give intrinsically better protection against cavitation than EG or PG.

It is our theory that some or all of these advantages are based upon the relative chelation ability of PDO versus EO and PO. The latter are readily able to chelate the ions. The chelate with EO and PO will be a five-membered ring which is relatively easy to form. PDO cannot chelate the ions as well because it forms a six-membered ring and this is more difficult.

EXAMPLES

Two chemistries were used in the following experiments. These are 1,3-propane diol (anhydrous) and 1,3-propane diol (50 to 85 percent volume percent aqueous solution).

Example 1

At the beginning, we believed that the classical corrosion and performance testing regimen as described in ASTM literature (2001 Annual Book of ASTM Standards, Volume 15. 05) provides an accepted method to evaluate and compare the corrosive properties of coolants to the metals customarily used in vehicle coolant systems. The new variable for fuel cells is the 400 volt (Direct Current) electric field and the issues that such a field presents to the coolant. Ionic inhibitors are

disqualified. The above coolants, running in the maximum resistance state with no inhibitors, were reviewed.

We believed that the following tests would accurately predict the above coolants' abilities to perform in a heat exchange system, in terms of corrosion protection, and physical and chemical properties. Since these new coolants had not been through this regimen of testing before, there was no experience or normal performance against which the tests could be compared for reasonableness. Therefore, each of the tests was controlled against 50 volume percent aqueous inhibited ethylene glycol.

The classical coolant development approach involves analyzing the fluid for physical and chemical properties. Once the properties are established, performance objectives are determined and the prototypes evaluated. These tests may be modified to better evaluate the performance of a coolant in its intended operating environment. Examples of modifications may include variations in the pressure, temperature, electric fuel environment, and duration of the tests. The data then will begin to serve to establish comparative and baseline data for the prototype new coolants. These tests will include fundamental properties, such as pH value and specific gravity, physical properties, and coolant-specific parameters including foaming tendency and reserve alkalinity. We believed that this data would direct the research towards the most appropriate coolants. The results are shown in Table 2.

30

Table 2: . Physical and Chemical Properties

Test Number & Description	Comparative Current Specification Value	Comments
ASTM D-1122 Relative Density <i>An experiment to determine the property of relative density. This information is used later in verifying the quality of commercialized products produced at blending facilities, and also has value to estimate contamination levels.</i>	1.110-1.145	The relative density of the new coolant will be different than EG or PG and will also depend on the concentration of PDO and water.
ASTM D-1177 Freeze Point <i>This experiment overcomes the soft 'slushy' freeze characteristic that makes determining the freezing point of some fluids difficult. It produces a graph of cooling behavior from which a consistent and meaningful freeze point can be determined.</i>	<-40°C	Choosing an appropriate solution can satisfy this requirement.
ASTM D-1120 Boiling Point <i>This is a boiling point method consistent with standard methods used to determine the boiling points of most fluids.</i>	>90°C	The boiling point of the new coolant will be different than EG or PG and will also depend on the concentration of PDO and water
ASTM D-1882 Auto Finish <i>The coolant is likely to be spilled on an auto finish. Therefore, it has always been a requirement that the coolant has no effect on the cars' finish, and this test was developed to evaluate that property.</i>	no effect	No problem expected.
ASTM D-1119 Ash Content <i>High levels of dissolved solids are associated with premature water pump wear and other durability issues. Completely evaporating the liquid and calculating the weight of the remaining dry material determines ash content.</i>	<5.0% max.	Since this coolant will be very low in inhibitors, this specification may need to be further reduced to prevent conductivity problems.
ASTM D-1287 pH: <i>The H⁺ ion concentration is reported as a pH value. This value is determined from an instrument reading. The pH value has to be appropriate for the inhibitor technology in use.</i>	7.5 to 11.0	Experimentation will likely result in a tighter spec for PDO than is used today for EG and PG coolants.
ASTM D-1123 Water mass percent <i>Water content on non-aqueous coolants is determined by the Karl Fischer method.</i>	5.0 %/max.	Applicable to the PDO before blending.

Test Number & Description	Comparative Current Specification Value	Comments
ASTM D-1121 Reserve Alkalinity <i>In many inhibition technologies, the durability of the coolant is related to its ability to neutralize weak acids formed as the base and/or inhibitors degrade. This titration evaluates that property.</i>		This property may be obsolete, or may have QC value.
ASTM D-1881 Foaming Tendencies <i>Foaming is an undesirable property associated with negative performance. This method creates a measurable volume, and also the time required to dissipate the foam.</i>	Break: 5 sec. Volume: 150 ml	The new coolant should meet this requirement.
Electrical Conductivity mohs Test method: a calibrated laboratory bench conductivity meter is employed to measure the conductivity of the coolant. The conductivity probe is placed into the fluid, and the digital reading on the conductivity meter is observed.	< 50	Experimental data to be used in developing a test and performance specification.
Viscosity (cPs) ASTM D-445	<1080 °C <600 °C	Comparable to EG coolant.
Thermal Conductivity W/m-K from literature	>0.4	Comparable to EG coolant.
Heat Capacity (kJ/kg-K) from literature	>3	Comparable to EG coolant.
Durability by extended duration tests	>5 years	PDO promises excellent stability.
Effect on Elastomers: By Cummins Method 14292 Silicon Seals, Viton, Bunan (Nitrile), Teflon, Neoprene, Rubber, Nylon	<10% Δ Dimension Each	

Test Number & Description	Comparative Current Specification Value	Comments
Toxicity LD ₅₀ data and review of MSDS	Non toxic for transportation.	PDO offers low toxicity.
ASTM D2809 Water Pump Test, repeated three times	≥8 each time	PDO has performed better than EG in the series of tests. See Table 3 below.
ASTM D-4340 Corrosion of Aluminum Heat Rejecting Surfaces	<1.0 mg/cm ² /week	PDO has performed at less than 10% of the allowed loss.
Extended aging evaluation in D-4340 Rig @ 150 °C for 60 Days sampled @ 10 day intervals. Aluminum weight loss Δ pH Oxidation products (i.e. COOH anions) Oxidation trend (slope of regression)	<1.0 mg/cm ² /week < 2 pH units <20% < 2,000 ppm m<1	PDO degraded less in terms of pH value and in the formation of oxidation by-products in the presence of two fully formulated coolant inhibition packages. See Table 4 below.
ASTM D-1384 Corrosion in Glassware (Higher Performance Specification) Copper Lead Solder Brass Steel Cast Iron Cast Aluminum	Maximum Weight Loss, mg 5 10 5 5 5 10	Test passed.
Aged Coolant Corrosion (ASTM D-1384 extended) in Glassware @ 150 °C (Fluid from 2,000 Hour Aging) Copper Lead Solder Brass Steel Cast Iron Cast Aluminum	Maximum Weight Loss, mg 10 30 10 10 10 30	
Erosion Corrosion of Heat Exchanger, 2,000 hours	No leaks	
Repassivation of Aluminum by Galvanostatic Measurement ASTM D6208	E _s < 2.0 E _g > -0.4.0	
ASTM D-2570 Simulated Service (Higher Performance Specification) Copper Lead Solder Brass Steel Cast Iron Cast Aluminum	Maximum Weight Loss, mg 10 20 10 10 10 20	Multiple embodiments passed.

Table 3

ASTM 2809 Test Data		
Inhibitor	EG	PDO
Conventional Automotive	8	9
Carboxylate Automotive	2	8
Phosphated Heavy Duty	10	10
Non Phosphated Heavy Duty	3	8
Hybrid Heavy Duty	9	10

Table 4

Oxidation comparison between PDO and EG inhibited with commercial inhibitor package @ 2.2%. Test run on D-4340 at 150°C, without corrosive water and at 50% concentration.							
			pH				
Time (days)	0	10	20	30	40	50	60
PDO-A	11.16	9.31	8.87	8.69	8.41	8.19	7.96
EG-A	10.06	7.67	6.38	5.68	4.60	4.31	4.07
PDO-B	10.58	9.63	8.89	8.56	8.32	8.18	7.93
EG-B	10.67	9.22	8.67	8.32	8.02	7.92	7.74
		Total Degradation Acids (ppm)					
Time (days)	0	10	20	30	40	50	60
PDO-A	0	213	415	607	762	851	1029
EG-A	0	542	1553	1987	3498	4028	4705
PDO-B	0	231	372	587	688	833	1053
EG-B	0	342	654	922	1128	1486	1602

5

Example 2

In these experiments, a solution of 50 percent by volume 1,3-propane diol (PDO) and 50 percent by volume deionized water were tested for corrosion of various metals used in engine cooling systems over a period of time. The test method was modified from ASTM test method D-2570 by using the spaced interval examination procedure detailed in ASTM G-31. The following Table 5 shows the results:

10

Table 5: Extended Spaced Interval Simulated Service Test
Modified from ASTM D2570 (using ASTM G31 spaced interval)
Test Method

- 5 PDO @ 50% in DI Water
190 °F (88°C). Spaced Interval Corrosion Data

Weeks	2	4	6	8	10
Copper	2	2	1	1	2
Lead Solder	3	2	6	6	3
Brass	2	2	3	3	4
Steel	11	12	13	13	13
Cast Iron	13	10	11	11	40
Cast Aluminum	22	34	40	40	40

- 10 Note how the corrosion behaves after 8-10 weeks. The fact that the aluminum corrosion does not increase after 6 weeks gives an indication that there is some flash corrosion initially but after that the oxides protect the aluminum. Generally, the absolute limit is specified by ASTM D3306 to be 60mg of aluminum lost after 7 weeks' exposure.

15 Example 3

The next experiment was corrosion of aluminum services over an extended period of time. The results are set out in Table 6 below.

- 20 Table 6: Corrosion of Heat Rejecting Aluminum Surface
Modified from ASTM D4340

Temperature elevated to 300 °F (149°C), Time extended from 1 week to 30 days

- 25 50% PDO 50% (volume) DI Water

	Before Test	10 Days	20 Days	30 Days
Weight loss mg/cm ² /week	-	0.0	0.0	0.0
pH	6.55	5.34	4.60	4.99
Conductivity µmhos/cm	0	9	9	.14
Comments	No damage to specimen	No damage to specimen	No damage to specimen	No damage to specimen

Please note that even after running this test for 30 days, there was no apparent corrosion damage to the specimen.

Example 4

5 This example describes experiments following the ASTM D1384 test method, modified by omitting the corrosive salts and were also made to operate at 150 degrees C by changing the bath from water to 50% propylene glycol. The tests were done to test the corrosivity of solutions of PDO in water
10 having amounts of PDO from 55 to 85 percent by weight. We have identified the 65 weight percent PDO solution as being the best because it offered the best overall protection for the six metals tested. However, the data in Table 7 also shows that solutions containing 55% and 60% PDO in water
15 also achieved very good results because fuel cell systems are most likely to be manufactured primarily of aluminum and stainless steel.

Table 7

Percent PDO in water	55	60	65	70	75	80	85
Copper	1.2	2.0	1.6	1.7	1.7	1.6	0.6
Lead Solder	123.8	93.5	62.5	60.3	39.2	63.7	20.3
Brass	2.1	1.7	1.8	2.0	1.7	2.7	1.2
Steel	126.1	86.8	84.6	15.8	29.2	26.3	1.5
Cast Iron	247.6	186.6	263	255.3	227.1	189.3	-0.7
Cast Aluminum	8.2	7.0	7.3	16.6	17.3	47.5	26.5
Conductivity Before Test $\mu\text{mhos/cm}$	0	0	0	0	0	0	0
Conductivity After Test $\mu\text{mhos/cm}$	30	22	10	7	4	3	0

20

Summary of Results

We believe that the results show that these PDO-based coolants can be used for a low conductivity application in

fuel cell powered systems, including fuel cell vehicles. PDO is demonstrated to be non-conductive and manifests corrosion resistant properties to the point of meriting serious consideration. The following are some of the more
5 significant findings:

- A coolant with high electrical resistance (low conductivity) has been developed that is appropriate for use in fuel cell powered systems, including fuel
10 cell powered vehicles, that generate strong electrical fields. It has electrical resistivity of more than 250 kohm-cm. Ethylene glycol is too corrosive to be completely nonconductive.
- The coolant, containing PDO, can be formulated in
15 various concentrations to achieve freeze points of -40 (°F or °C) or lower (see freeze point graphs in Figures 1 and 2).
- The coolant offers more favorable boiling points in aqueous solutions than traditional glycol based
20 coolants, as high as 471 °F (234°C).
- The thermal conductivity is comparable to glycol-based coolants (in water).
- The viscosity is comparable to glycol-based coolants (in water).
- 25 • The heat capacity is comparable to glycol-based coolants (in water).
- The durability is better than glycol based coolants, offering the prospect of a closed, lifetime-filled low or no maintenance coolant system.
- 30 • The coolant is compatible with system materials, including aluminum and elastomers.

- The coolant is less toxic and less palatable than ethylene glycol and is much less likely to be involved in pet or child poisonings.

- The cost of the coolant over the life of the system is comparable to existing premium coolants.

The physical property data for PDO and potentially competing coolants, ethylene glycol (EG) and propylene glycol (PG) are shown in Table 8:

Table 8

Physical Properties	PDO	EG	PG
Mol. Wt.	76.1	62.07	76.1
Boiling Point, °F (°C)	417.9 (214.4)	387.7 (197.6)	369.3 (187.4)
Flash Point, °F (°C)	265 (129)	240 (116)	220 (104)
Specific Gravity, 20°C	1.0526	1.115	1.032
Freeze Point, 50% solution, °F (°C)	-21 (-29)	-36 (-38)	-28 (-33)
Pour Point, °F (°C)		<-75 (<-59)	<-71 (<-57)
Viscosity, cP 20°C	52	17	49
Specific Heat, 212°F BTU/lb/F [kJ/(kg*K)]	0.652 (2.730)	0.665 (2.784)	0.704 (2.948)
Thermal Conductivity, 25°C BTU/hr-ft-F [W/(m*K) @ 25°C]	0.127 (0.220)	0.147 (0.254)	0.119 (0.206)
Heat of Vaporization 25°C, BTU/lb [kJ/kg @ 25°C]	410 (954)	449 (1044)	379 (882)
Purity	99.7	94.5	99

WE CLAIM:

1. A nontoxic fuel cell engine coolant which has an electrical resistivity of greater than 250 kOhm-cm, a boiling point of greater than 90°C, a thermal conductivity of greater than 0.4 W/m-k, a viscosity of less than 1 cPs at 80°C, a viscosity of less than 6 cPs at 0°C, a heat capacity of greater than 3 kJ/kg-K, and which is compatible with current cooling system materials.

2. The coolant of claim 1 wherein the coolant is 1,3-propanediol.

3. The coolant of claim 1 is an aqueous solution comprised of from 1 to 100% by volume of 1,3-propanediol.

4. The coolant of claim 3 wherein the solution is comprised of from 40 to 85% by volume of 1,3-propanediol.

5. The coolant of claim 4 wherein the solution is comprised of from 55 to 85% by volume of 1,3-propanediol.

6. The coolant of claim 1 having a freezing point of less than -40°C.

Exhibit 2

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technical advice/conductivity

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Measurement of Conductivity

Conductivity Measurement Theory and Practice

Background

Electrical conductivity is an inherent property of most materials, and ranges from extremely conductive materials like metals to very non-conductive materials like plastics or glass. About halfway between the two extremes in conductivity are aqueous solutions, such as sea water and plating baths.

In metals, the electrical current is carried by electrons, while in water it is carried by charged ions. In both cases, the conductivity is determined by the number of charge carriers, how fast they move, and how much charge each one carries.

Thus, for most water solutions, the higher the concentration of dissolved salts, which will lead to more ions, the higher the conductivity. This effect continues until the solution gets "too crowded," restricting the freedom of the ions to move, and the conductivity may actually decrease with increasing concentration. (This can result in two different concentrations of a salt having the same conductivity.) See **Table 1**.

Some species ionize more completely in water than others do, and their solutions are more conductive as a result. Each acid, base, or salt has its own characteristic curve for concentration vs. conductivity.

Metals are extremely conductive because electrons move almost with the speed of light, while in water ions move much slowly, and the conductivity is much lower. Raising the temperature makes water less viscous, and the ions can move faster. Because the ions are of different sizes, and carry different amounts of water with them as they move, the temperature effect is different for each ion. Typically, the conductivity varies about 1-3% per degree C, and this temperature coefficient may itself vary with concentration and temperature. See **Tables 1 and 2**.

Definitions

The conductivity of a material is an inherent property-that is, pure water at a particular temperature will always have the same conductivity. The *conductance* of a sample of pure water depends on how the measurement is made-how big a sample, how far apart the measuring electrodes are, etc. It is defined as the reciprocal of the resistance in ohms, measured between the

opposing faces of a 1 cm cube of liquid at a specific temperature. See **Figure 1**. The basic unit of conductance is the *Siemens* (S) and was formerly called the mho. Because a measurement gives the conductance, techniques have been worked out to convert the measured value to the conductivity, so that results can be compared from different experiments. This is done by measuring a *cell constant* for each setup, using a solution of known conductivity.

Cell Conductance X K = Conductivity (Equation 1)

The cell constant is related to the physical characteristics of the measuring cell. K is defined for two flat, parallel measuring electrodes as the electrode separation distance (d) divided by the electrode area (A). Thus, for a 1 cm cube of liquid,

$$K = d/A = 1 \text{ cm}^{-1} \text{ (Equation 2)}$$

In practice, the measured cell value is entered into the meter, and the conversion from conductance to conductivity is done automatically. The K value used varies with the linear measuring range of the cell selected. Typically, a cell with $K = 0.1 \text{ cm}^{-1}$ is chosen for pure water measurements, while for environmental water and industrial solutions a cell with K of 0.4 to 1 cm^{-1} is used. Cells with up to $K = 10 \text{ cm}^{-1}$ are best for very high conductivity samples.

For some solutions, such as pure water, the conductivity numbers are so low that some users prefer to use *resistivity and resistance* instead. The resistivity is the reciprocal of the conductivity ($R = 1/C$), and the resistance is the reciprocal of the conductance. Resistance units are in ohms, and 1 ohm = 1/Siemens. From Eq. 1 and 2, it can be seen that conductivity units are in Siemens/cm, and therefore resistivity units are in ohm-cm. measured temperature, temperature coefficient and reference temperature, will report sample conductivity

How is conductivity measured?

In the simplest arrangement (a *2-electrode* cell), a voltage is applied to two flat plates immersed in the solution, and the resulting current is measured. See **Figure 1**. From Ohm's Law, the conductance = current/voltage. Actually there are many practical difficulties. Solution conductivity is due to ion mobility. Use of DC voltage would soon deplete the ions near the plates, causing *polarization*, and a higher than actual resistance. This can be mostly overcome by using AC voltage, but then the instrument designer must correct for various capacitance and other effects. Modern sophisticated 2-electrode conductivity instruments use complex AC waveforms to minimize these effects, and by using the cell constant, measured temperature, temperature coefficient and the reference temperature will report sample conductivity.

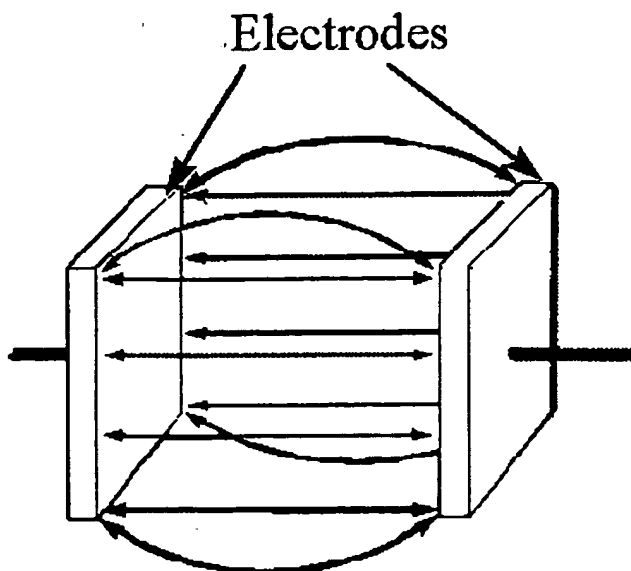


Figure 1.
Conductivity Cell

Table 1
Some Conductivity Values of Typical Samples

Sample at 25°C	Conductivity microScm ⁻¹
Ultrapure Water	0.055
Power Plant Boiler Water	1.0
Drinking Water	50
Ocean Water	53,000
5% NaCl	223,000
50% NaOH	150,000
10% HCl	700,000
32% HCl	700,000
31% HNO ₃	865,000

Table 2
Some typical temperature coefficients

Sample	Percent per °C (at 25°C)
Ultrapure Water	4.55
Salt Solution (5%)	2.12
NaOH (5%)	1.72
Dilute Ammonia Solution	1.88
HCl (10%)	1.32
Sulphuric Acid (5%)	0.96
Sulphuric Acid (98%)	2.84

Sugar Syrup	5.64
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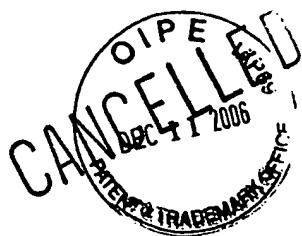
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Exhibit 3



Patent
TH1871Y (US)
RBT

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re application of)

EDWARD R. EATON ;)
WYNDHAM H. BOON; and)
CHRISTOPHER J. SMITH)

Serial No. 10/074,834)

Group Art Unit: 1751

Filed February 13, 2002)

Examiner: Ogden Jr., N.

A NOVEL CHEMICAL BASE FOR FUEL)
CELL ENGINE HEAT EXCHANGE)
COOLANT/ANTIFREEZE)

COMMISSIONER FOR PATENTS
P. O. Box 1450
Alexandria, VA 22313-1450

Sir:

DECLARATION UNDER 37 CFR §1.132

Glenn Komplin declares as follows:

1. I am employed by Shell Global Solutions (US) Inc. as a Staff Research Engineer.
2. I received a Bachelor of Science in Chemical Engineering from the University of Wisconsin, Madison in 1984, and a Ph.D in Material Science from the University of Wisconsin, Madison in 1990.
3. I performed an experiment designed to measure the conductivity of aqueous solutions of 1,3-propanediol and deionized water. I prepared 3 solutions of 1,3-

propanediol/deionized water—1) a solution of 80 wt.% 1,3-propanediol and 20 wt.% deionized water; 2) a solution of 50 wt.% 1,3-propanediol and 50 wt.% deionized water; and; 3) a solution of 20 wt.% 1,3-propanediol and 80 wt.% deionized water. I then measured the conductivity of the three solutions at 24°C with an Orion Model 115 conductivity meter. The results are shown in Table 1 below where 1,3-propanediol is denoted as PDO and deionized water is denoted as DI water.

TABLE 1
(Conductivity of 1,3-propanediol/deionized water solutions in μ Seimens/cm)

80% PDO/20% DI water	50% PDO/50% DI water	20% PDO/80% DI water
0.8	2.2	4.3

4. I performed an experiment designed to measure the conductivity of aqueous solutions of 1,3-propanediol and tap water. I prepared 3 solutions of 1,3-propanediol/tap water—1) a solution of 80 wt.% 1,3-propanediol and 20 wt.% tap water; 2) a solution of 50 wt.% 1,3-propanediol and 50 wt.% tap water; and; 3) a solution of 20 wt.% 1,3-propanediol and 80 wt.% tap water. I then measured the conductivity of the three solutions at 24°C with an Orion Model 115 conductivity meter. The results are shown in Table 2 below where 1,3-propanediol is denoted as PDO.

TABLE 2
(Conductivity of 1,3-propanediol/tap water solutions in μ Seimens/cm)

80% PDO/20% tap water	50% PDO/50% tap water	20% PDO/80% tap water
9.2	72.5	273

5. The electrical resistivity of the 1,3-propanediol/deionized water solutions and the 1,3-propanediol/tap water solutions in ohms-cm may be calculated by dividing 1 by the conductivity of these solutions. The electrical resistivity in Kohm-cm may be calculated from the electrical resistivity in ohm-cm by dividing the electrical resistivity in ohm-cm by 1000. The calculated electrical resistivity of the 1,3-propanediol/deionized water solutions and the 1,3-propanediol/tap water solutions in Koh-cm is shown in Table 3 below, where 1,3-propanediol is denoted as PDO and deionized water is denoted as DI water.

TABLE 3
(Electrical resistivity of 1,3-propanediol water solutions in Kohm-cm)

	80%PDO/20% water	50%PDO/50% water	20%PDO/80%water
DI water	1250	455	232
Tap water	108	14	4

Glen C Kopp

Date: December 4, 2006

Exhibit 4



US 20050013753A1

(19) **United States**(12) **Patent Application Publication** (10) **Pub. No.: US 2005/0013753 A1****Eaton et al.**(43) **Pub. Date: Jan. 20, 2005**(54) **HEAT-TRANSFER SYSTEMS****Publication Classification**(76) Inventors: **Edward Eaton**, Phoenix, AZ (US);
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(US)(51) **Int. Cl.⁷** **F28D 1/00**
(52) **U.S. Cl.** **422/198; 422/200**Correspondence Address:
STONEMAN LAW OFFICES, LTD
3113 NORTH 3RD STREET
PHOENIX, AZ 85012 (US)(57) **ABSTRACT**(21) Appl. No.: **10/886,298**(22) Filed: **Jul. 6, 2004****Related U.S. Application Data**(60) Provisional application No. 60/485,441, filed on Jul.
7, 2003.

An improved system for transferring heat, utilizing diols, specifically 1,3-propanediol (PDO). The system reduces the energy consumption, maintenance costs, corrosion, toxicity, and the risk of fire associated with such heat-dependent processes. Unlike traditional hydrocarbon heating fluids, diols do not require draining and replacement if a purifier is installed in the system. Business methods are also disclosed.

